Int J Life Cycle Assess (2014) 19:307–319
DOI 10.1007/s11367-013-0625-x

LCA OF WASTE MANAGEMENT SYSTEMS

Design of recycling system for poly(methyl methacrylate) (PMMA). Part 2: process hazards and material flow analysis

Yasunori Kikuchi · Masahiko Hirao · Hirokazu Sugiyama · Stavros Papadokonstantakis · Konrad Hungerbühler · Takashi Ookubo · Akinobu Sasaki

Received: 17 October 2012 / Accepted: 5 July 2013 / Published online: 24 July 2013

Abstract
Introduction In this series of papers, we present a design of poly(methyl methacrylate) (PMMA) recycling system considering environmental impacts, chemical hazards, and resource availability. We applied life cycle assessment (LCA), environment, health, and safety (EHS) assessment as well as material flow analysis to the evaluation of the recycling system.
Purpose Recycling systems for highly functional plastics such as PMMA have not been studied sufficiently. Along with the popularization of PMMA-containing products such as liquid crystal displays (LCDs), the use of PMMA is steadily increasing, which will result in more waste of PMMA in the next decades. In this study, pyrolysis process for recycling waste PMMA into methyl methacrylate (MMA) monomer was examined, considering not only general environmental impacts quantified by life cycle assessment but also local environment, health, and safety hazards, and raw material availability.
Methods Process EHS hazards assessment was applied to quantify the local effects of the PMMA monomer recycling process. Process hazards are strongly connected with the hazardous properties of chemical substances and stream conditions within the process. Two alternative cooling methods exist, and their difference was analyzed by LCA and EHS assessment. Besides the process hazard, the availability of waste PMMA must be an important point for the feasibility of implementing the PMMA monomer recycling process. The available amount can be quantified by analyzing the material flow of PMMA-containing products. PMMA contained in LCDs as light guide panels was selected as a feasible source of waste PMMA, and the quantity of PMMA flows in the society was evaluated.
Results and discussion In the case of PMMA, monomer recycling has less process hazard than the production of fresh MMA from crude oil. The implementation of circulated cooling water could significantly decrease the process hazard in PMMA pyrolysis attributable to chemical hazards. Material flow analysis revealed that the availability of waste PMMA shows a fluctuating trend in the next 20 years because of the sharp peak demand for LCD television sets. The fluctuation is strongly dependent on the lifetime of LCD television sets.
Conclusions PMMA monomer recycling has a potential to reduce environmental impacts with a less process hazards than

Responsible editor: Yasunari Matsuno

Electronic supplementary material The online version of this article (doi:10.1007/s11367-013-0625-x) contains supplementary material, which is available to authorized users.

Y. Kikuchi (✉)
Presidential Endowed Chair for "Platinum Society",
The University of Tokyo, Ito International Research Center,
7-3-1 Hongo, Bunkyo-ku 113-0033, Tokyo, Japan
e-mail: kikuchi@platinum.u-tokyo.ac.jp

Y. Kikuchi · M. Hirao
Department of Chemical System Engineering, the University of Tokyo, 7-3-1 Hongo, Bunkyo-ku 113-8656, Tokyo, Japan

M. Hirao
e-mail: hirao@chemsys.t.u-tokyo.ac.jp

H. Sugiyama · S. Papadokonstantakis · K. Hungerbühler
Institute for Chemical- and Bioengineering, Safety and Environmental Technology Group, Swiss Federal Institute of Technology Zurich (ETHZ), Wolfgang-Pauli-Strasse 10,
8093 Zurich, Switzerland

H. Sugiyama
e-mail: sugiyama@chemsys.t.u-tokyo.ac.jp

S. Papadokonstantakis
e-mail: stavros.papadokonstantakis@chem.ethz.ch

K. Hungerbühler
e-mail: konrad.hungerbuehler@chem.ethz.ch

T. Ookubo · A. Sasaki
Mitsubishi Rayon, co., Ltd, 1-1, Marunouchi 1-Chome,
Chiyoda-ku, Tokyo 100-8253, Japan

T. Ookubo
e-mail: ookubo_ta@mrc.co.jp

A. Sasaki
e-mail: sasaki_ak@mrc.co.jp

fresh MMA production from crude oil. The availability of waste PMMA has a strong relationship with the lifetime of LCD television sets. The multiple and comprehensive assessments can reveal various aspects of a process technology.

Keywords EHS hazard · Liquid crystal display · Material flow analysis · PMMA pyrolysis · Weibull distribution

1 Introduction

The recycling of plastics has become an important issue all over the world. Some systematic analyses on recycling systems have been conducted for solid waste management using life cycle assessment (LCA) (Arena et al. 2003a, 2003b), waste management of soft drink packaging systems in Mexico (Romero-Hernández et al. 2009), and the transboundary recycling of polyethylene terephthalate (PET) bottles between Japan and China (Nakatani et al. 2010). In this regard, however, a process should be designed and analyzed in detail to bring a new recycling method into practice. When we develop a process for sustainability, multiple aspects must be carefully analyzed and interpreted for practical implementation. Environmental impacts quantified by LCA are one such aspect and should be interpreted with other aspects assessed by other tools. Such a multiple assessment approach has been discussed for process systems design (Kikuchi and Hirao 2009) and within green engineering (Jiménez-González and Constable 2011). In part 1 of this series of papers, it was demonstrated that the environmental impacts originating from the life cycle of a unit amount of PMMA can be reduced by a pyrolysis process, converting waste PMMA into recycled MMA monomer (Kikuchi et al. 2013, accepted). Through the investigation on a plant for collecting, sorting, and processing of electronic devices containing PMMA, existing recycling systems based on recycling laws and regulations are applicable as a part of PMMA recycling system (Kikuchi et al. 2013, accepted). Note that recycling system includes collection, logistics, and other processes as well as recycling process.

In the actual process design, local risks such as environment, health, and safety (EHS) issues are inevitable aspects and, generally, have higher priority than global environmental impacts, where local environmental risk mainly means the local and accidental effects of the release of hazardous chemicals (Kolluru et al. 1996). Process safety has been regarded as the most important issue in process design and operation. In the life cycle stages of process development, various technical issues on process safety must be addressed (CCPS 2007). The PMMA monomer recycling process is based on pyrolysis of PMMA resin and may be operated at relatively high temperature. During the heat decomposition of PMMA, chemical substances with a small molecular size, which may have high EHS hazards such as low flash point or high mobility, can be generated as by-products (Kaminsky and Franck 1991; Scheirs and Kaminsky 2006). To treat such by-products adequately, additional devices and energy may be required, and trade-off relationships may exist among evaluation indicators (Kikuchi and Hirao 2009).

The feasibility factors of processes such as aspects of economic or resource availability must also be carefully studied because a process with unfeasible factors may be rejected, even if they have outstanding characteristics, e.g., high profits or low environmental impacts. For a recycling process, the availability and quality of collected wastes, i.e., raw materials, are strongly related to its feasibility because raw materials for recycling process have a more dynamic availability profile, e.g., compared to fossil resources as recognized in PET recycling case in Japan (Nakatani et al. 2010). The forecasting of waste availability can be important information for specifying the most effective plant scale to be constructed. Although a large process throughput can increase the efficiency of processes such as heating or cooling and the profit through economic scale factors, it cannot be sustainable without a consideration of available raw materials. With respect to the PMMA monomer recycling process, the throughput should be based on the future availability of collectable waste PMMA because it is a continuous process and will have to operate over decades. According to statistical and market forecasting reports, the demand for PMMA resin will keep increasing because of the widespread use of liquid crystal display (LCD) panels for television sets (Display Search Ltd 2012). A recycling system of PMMA is strongly needed for the waste PMMA due to such an increasing demand for LCD.

In this study, we aim to design a PMMA recycling system with a PMMA monomer recycling process. In this series of papers, we assess the acceptability of a PMMA recycling process as a social technology by an evaluation of the different aspects of the PMMA monomer recycling process. The LCA of the PMMA monomer recycling scenarios was conducted in part 1 (Kikuchi et al. 2013, accepted). In this paper, we conducted assessments of two more aspects of the PMMA recycling process and system towards their practical implementation. First, process hazards of the PMMA monomer recycling were evaluated with an analysis of the relationship between local EHS risks and environmental impacts quantified by LCA. Second, the future availability of the raw material, i.e., waste PMMA, was analyzed by material flow analysis (MFA). As for the target products, LCD products (television sets, laptops, and monitors) were selected because of the possibility of their collection based on existing recycling systems. Based on the results of these assessments, the points to be addressed in a practical implementation of PMMA monomer recycling were extracted and discussed.

2 Materials and methods

2.1 Process design framework with multiple assessment methods

Figure 1 shows a proposed process assessment framework for a comprehensive analysis, where global environmental impact, local chemical risks, and feasibility of a process are concurrently assessed and comprehensively interpreted (Kikuchi and Hirao 2009). Global environmental impacts originating from PMMA monomer recycling were examined in part 1 of this series of papers. Other aspects are studied in this paper, as indicated by process hazards and resource availability for local risks and feasibility. In this study, ETH–EHS assessment and MFA originating from Japanese demand for PMMA products were selected for their quantification. The methods for individual assessments are described below.

2.2 Process hazard assessment

2.2.1 Background

Based on the life cycle of process development, various safety assessment methods have been developed and systematically compared (Adu et al. 2008; Koller et al. 2001) and can be mainly separated into two groups: hazard-based and detailed methods (Koller et al. 2000; Kolluru et al. 1996). Detailed methods require individual machine information, plant structure, and actual operation (Kolluru et al. 1996). Because such methods cannot be readily applied in the design phase without highly uncertain assumptions, hazard-based methods have been widely used in early the design phase. Among the quantitative EHS assessment methods, the ETH–EHS method (Koller et al. 2000) is one of the methods considering three EHS categories (Adu et al. 2008). It can take into account not only chemical hazards but also process conditions such as stream temperature and compositions. Using this method, various MMA production routes from crude oil (Sugiyama et al. 2009) were assessed by the ETH–EHS method (Sugiyama et al. 2008b). PMMA pyrolysis produces MMA monomer from waste PMMA. Because recycled MMA monomer has physical properties comparable with fresh MMA from crude oil, the PMMA pyrolysis process can be regarded as MMA production process. By applying the ETH–EHS method as the assessment method for quantifying local risks of PMMA pyrolysis in this study, the process hazards of PMMA monomer recycling are compared with those of fresh MMA production from fossil resources, which were evaluated by Sugiyama et al. (2008b) using Aspen Plus simulator. Note that the ETH–EHS method quantifies process hazard attributable to chemical hazards and does not include detail process risks occurred by the probabilistic factors such as malfunction, aging deterioration of equipment, or human errors.

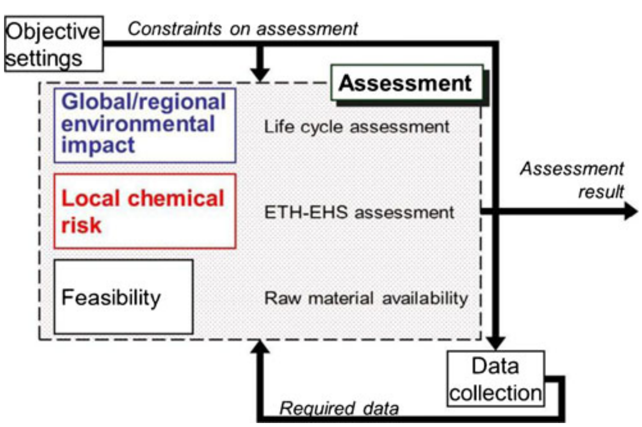


Fig. 1 Outline of multiple process assessments based on a risk-based decision framework (Kikuchi and Hirao 2009)

2.2.2 Method

The ETH–EHS method originally developed by Koller et al. (2000) has been modified to quantify process hazards originating not only from hazardous or physical properties of substances but also from process characteristics. In this study, the method proposed by Sugiyama (2007, 2008b) was adopted to correspond to the same conditions as those used for the assessment of fresh MMA production processes. The equations in this method (Sugiyama et al. 2008b) are the following:

$$\mathrm{SH} = \sum_{\mathrm{cS}} \max_{F} \left(\sum_{i} m_i^F \cdot \mathrm{I}_i^{\mathrm{cS}} \right), \tag{1}$$

$$\mathrm{HH} = \sum_{\mathrm{cH}} \max_{i} \left(\sum_{i} m_i^{\mathrm{UN}} \cdot \mathrm{I}_i^{\mathrm{cH}} \right), \tag{2}$$

$$\mathrm{EH} = \sum_{\mathrm{cE}} \sum_{i} \left(z \cdot \max_{F} \left(m_i^F \right) \cdot \mathrm{I}_i^{\mathrm{cE}} \right) + \sum_{\mathrm{cE}} \sum_{j} m_j^{\mathrm{Out}} \cdot \mathrm{I}_j^{\mathrm{cE}}, \tag{3}$$

Where SH/HH/EH is safety/health/environment hazard (in kilograms per kilogram-product); cS/cH/cE is the number of safety/health/environment hazard categories (acute toxicity, reaction/decomposition, fire/explosion, and mobility for safety, chronic toxicity, and irritation for health, solid waste, water hazard, air hazard, and persistency in the environment); F is flow in process; i is chemical substances; m is mass per unit amount of product (in kilograms per kilogram-product); $\mathrm{I}_i^{\mathrm{cS}}/\mathrm{I}_i^{\mathrm{cH}}/\mathrm{I}_i^{\mathrm{cE}}$ is the index value of chemical substance i in safety/health/environment hazard category cS/cH/cE [−]; m^{UN} is the unit amount of mass, 1 kg; z is

the emission factor of chemical substances to the environment, 0.1 in this study corresponding with existing literature (Sugiyama et al. 2008b); and m^{Out} is the amount of directly emitted chemical substances from the site to the environment (in kilograms per kilogram-product).

2.2.3 Evaluation settings

Based on Eqs. (1) to (3), process hazards are quantified. The boundary of this process hazard assessment is the gate-to-gate of the PMMA pyrolysis process, as shown in Fig. 2a. The conditions for each flow, such as the compositions of chemical substances, flow rate, temperature, and pressure, were obtained from an actual pilot plant. To calculate the index values for chemical substances in each category, the databases of hazardous and physical properties of chemical substances were applied (CDC 2012; ChemExper Inc 2012; EPA 2011; ILO 2012; Sugiyama 2007). According to the investigation of the actual pilot plant, the emission during steady-state operation indicated in the second term of Eq. (3) can be neglected because all of the flows except the product stream in Fig. 2a are connected to the heat recovery system.

Two process alternatives exist regarding the process flow in PMMA pyrolysis as organized in Table 1. In the process flow diagram of PMMA pyrolysis shown in Fig. 2a, there is a condenser unit where gaseous substances are condensed by cooling the effluent gas from the pyrolysis reactor. In this unit, the effluent can be cooled by a recycling flow or cooling water, as shown in Fig. 2a, b, respectively. For this cooling, high flows of recycled flow and cooling water are required. On the process hazard assessment, flows with a larger amount of hazardous material can result in a higher hazard, while the use of a greater amount of cooling water may cause higher environmental impacts. In this study, the changes in process hazards and environmental impacts are analyzed by the ETH–EHS and LCA assessment methods, respectively. Existing LCA databases were utilized for collecting of background data in Japan (JEMAI 2007; JLCA 2012).

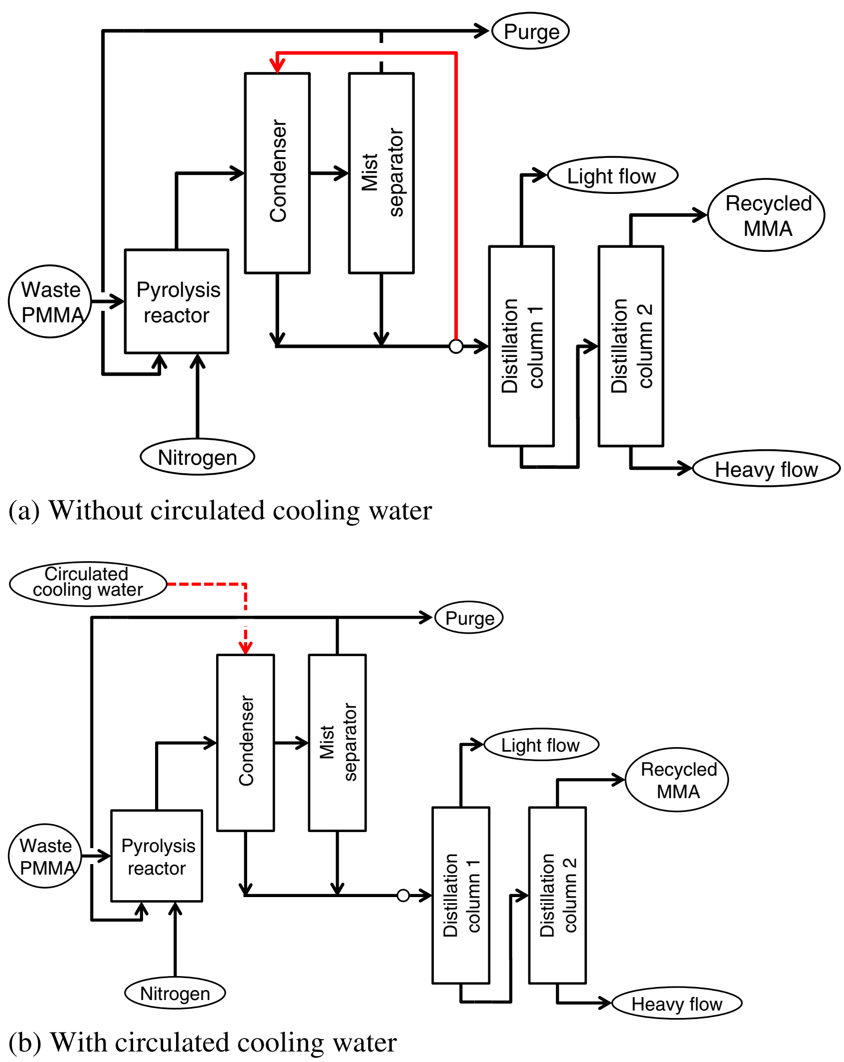


Fig. 2 Boundary of process hazard assessment of PMMA monomer recycling

Table 1 Scenario settings in two assessments. Scenarios 1 to 4 are the settings in case study 1, and scenario 5 shows the settings in case study 2

Assessment	Scenario	Settings
Process hazard	Flow use	Utilization of process flow as coolant
	Water use	Utilization of circulated cooling water
Availability of raw material	TV scenario 1	The status quo in Weibull parameters of television sets
	TV scenario 2	The same Weibull parameters for LCD television sets as CRT sets in 2010
	TV scenario 3	The same Weibull parameters for LCD television sets as CRT sets in 2015

2.3 Material flow analysis

2.3.1 Background

MFA has been utilized for analyzing the flows of target materials within a boundary (Lin 2008) and was applied for predicting waste generation (Liu et al. 2006) The PMMA products considered in this study were those deemed highly collectable after use in Japan. Considering existing recycling law (METI 2001; PC3R Promotion Association, Japan 2004) and existing recycling plants (Hirasawa 1999), LCD panels as a component of television sets, laptop computers, and monitors were regarded as the most collectable PMMA products and set as the target PMMA products in this study. LCD panels have a light guide panel (LGP) to ensure that light-emitting diode (LED) fluorescent light is uniformly dispersed. According to the literature on plastic products, nearly 100 % of LCDs used in Japan have been made from PMMA (Fuji Chimera Research Institute, Inc 2010). The production amount of television sets was drastically changed from 2009 to 2012 in Japan because of the switch over to terrestrial digital broadcasting implemented on July 24, 2011 (MIAC 2011). Because a data converter is needed for a television set to receive digital broadcasting, older types of television sets have been mostly replaced by new types. A special demand under this rare situation occurred from 2009 to the first half of 2011. The shipment of television sets was considerably reduced in the latter half of 2011 (JEITA 2012) and may maintain a low level during 2012 (JEITA 2011). In this paper, an analysis of material flow of PMMA contained in LCD products was conducted. While some parts of post-consumer LCD products may be exported from Japan, LCD products targeted in this study are regulated to recycling as shown above, which means that little parts of them may be exported.

2.3.2 Method

To analyze the material flows of PMMA contained in LCD products, shipment of products, contained mass per LCD products, and annual collectable amount of waste products must be forecasted. The scope of the MFA is shown in Fig. 3. The scope is a part of the life cycle of PMMA, as shown in Fig. 3a. As a detailed scope shown in Fig. 3b, it is assumed that the LCD panels containing LGP of categories A and B cannot be distinguished after LCD panel production. The material flows of PMMA contained in LCD products can be analyzed on the basis of those LCD products. Because the use period of LCD products is not 1 year, the balance between population and product stock in markets should be solved with the shipments and wastes of products (Yokota et al. 2003; Kakudate et al. 2002):

$$\begin{aligned}\mathrm{Market}_i^{\mathrm{LCD}}(t) &= \mathrm{P}_i^{\mathrm{LCD}}(t) - \mathrm{W}_i^{\mathrm{LCD}}(t) + \mathrm{Market}_i^{\mathrm{LCD}}(t-1)\\ &= \frac{\mathrm{Population}(t)}{\mathrm{Population}(t-1)}\cdot\frac{\mathrm{OR}_i^{\mathrm{LCD}}(t)}{\mathrm{OR}_i^{\mathrm{LCD}}(t-1)}\cdot\mathrm{Market}_i^{\mathrm{LCD}}(t-1),\end{aligned} \tag{4}$$

therefore,

$$\begin{aligned}\mathrm{P}_i^{\mathrm{LCD}}(t) = &\left(\frac{\mathrm{Population}(t)}{\mathrm{Population}(t-1)}\cdot\frac{\mathrm{OR}_i^{\mathrm{LCD}}(t)}{\mathrm{OR}_i^{\mathrm{LCD}}(t-1)}-1\right)\cdot\mathrm{Market}_i^{\mathrm{LCD}}(t-1)\\ &+\mathrm{W}_i^{\mathrm{LCD}}(t),\end{aligned} \tag{5}$$

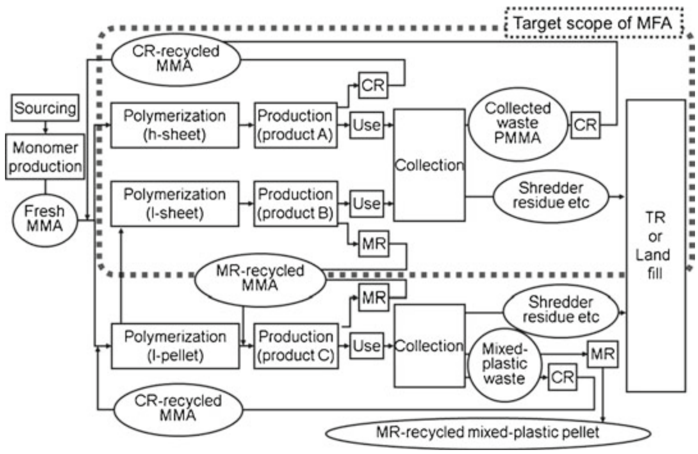
where

$$\mathrm{OR}_i^{\mathrm{LCD}}(t+1) = \mathrm{a}_i^{\mathrm{LCD}}(t)\cdot\mathrm{OR}_i^{\mathrm{LCD}}(t), \tag{6}$$

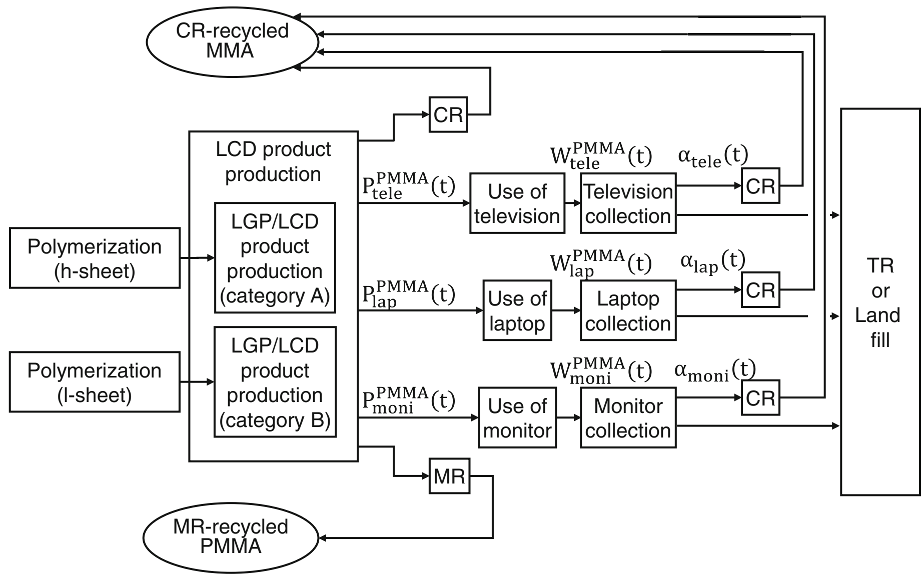
$$\mathrm{a}_i^{\mathrm{LCD}}(t) = \begin{cases} \mathrm{b}\cdot t + \mathrm{c} & \left(\mathrm{OR}_i^{\mathrm{LCD}}(t) < 1\right)\\ 1 & \left(\mathrm{OR}_i^{\mathrm{LCD}}(t) = 1\right)\end{cases} \tag{7}$$

Equation (4) shows the relationship among the market stock of LCD product i at year t ($\mathrm{Market}_i^{\mathrm{LCD}}(t)$ (set-product per year), shipment of LCD product i at year t ($\mathrm{P}_i^{\mathrm{LCD}}(t)$ (set-product per year), and waste of LCD product i at year t ($\mathrm{W}_i^{\mathrm{LCD}}(t)$ (set-product per year), which can also be calculated as the growth of market for LCD product i as described by the relationship between population at year t (population(t) [person]) and the ownership ratio of LCD product i at year t ($\mathrm{OR}_i^{\mathrm{LCD}}(t)$ (set-product per person) defined in Eqs. (6 and 7). Equation (7) means that the growth rate of ownership is assumed not to decrease. Hence, Eq. (5) can be used to predict the shipment of LCD products and required parameters for MFA in this study. Note that $\mathrm{P}_i^{\mathrm{LCD}}(t)$ and $\mathrm{W}_i^{\mathrm{LCD}}(t)$ are the total shipment and waste from the beginning to the end of year t. It is assumed that the shipped products at year t are not released as waste within the year.

Fig. 3 Description of the scope of MFA in this study as a part of the life cycle of PMMA discussed in part 1 of this series of papers (Kikuchi et al. 2013 submitted part 1), where $P_i^{PMMA}(t)$ is the amount of PMMA contained in shipped LCD product i at year t (kg y^{-1}), $W_i^{PMMA}(t)$ is the amount of PMMA contained in waste LCD product i at year t (kg y^{-1}), and α_i (t) is the ratio of waste PMMA sent to the monomer recycling process



(a) Target scope of MFA within the life cycle of PMMA based on Part 1 (Kikuchi et al. submitted Part 1)



(b) Scope of MFA

Equation (8) gives the way to calculate $P_i^{PMMA}(t)$ from $P_i^{LCD}(t)$:

$$P_i^{PMMA}(t) = \rho^{PMMA} \sum_l \frac{L_l^{LCD}}{\left(^{L}/_{th}\right)_{thin-wall}(t)} \cdot S_l^{LCD} \cdot r_{i,l} \cdot P_i^{LCD}(t), \tag{8}$$

Where $r_{i,l}$ is the ratio of screen size type l in LCD product i shipped at year t (–); S_l^{LCD} is the area of screen size type l (in square meters); L_l^{LCD} is the length of the short side of LCD screen size type l (in meters); and ρ^{PMMA} is the average density of PMMA resin [10^3 kg m^{-3}]. S_l^{LCD} and ρ^{PMMA} are constants for LCD products on the basis of industrial standards. Values of $r_{i,l}$ for the past shipments of LCD product i were based on statistical records (JEITA 2012). To calculate the contained PMMA as LGP in LCD products, the thickness of LGP should be estimated, which has a strong correlation with the level of thin-wall processing technology (Kubo 2011). The level for LGP can be indicated by the factor of flow length per thickness at year t ($\left(^{L}/_{th}\right)_{thin-wall}(t)$ [meter flow length (meter thickness)$^{-1}$]), and the state-of-the-art technology level can be obtained from the roadmap of thin-wall processing machines (Kubo 2011). From the literature on thin-wall processing technology (Kubo 2011), the coefficient of the top-technology level from 2000 to 2011 was extracted, and the average level was estimated by using the record of the used amount of PMMA for LGP in 2009 in Japan (Fuji Chimera Research Institute, Inc 2010) (see also Electronic Supplementary Material). Based on this approximation model, the thickness was calculated for the products shipped in 2010 and 2011, and it was confirmed that the thickness estimated by this model has adequate agreement with the actual shipment.

The number of discarded electric appliances was assumed to be correlated with product age and failure ratio in this study. The age was estimated by investigating the average of used products. It was assumed that the failure rate is determined on the basis of a statistical distribution function. Several functions have been applied for the material flow analyses in LCA research fields as follows: triangular and rectangular functions for the effects of household appliance recycling (Nakano et al. 2007), gamma function for air conditioners (Yokota et al. 2003) and passenger cars (Kakudate et al. 2002), and Weibull for the prediction of the service life of products (Aktas and Bilec 2012). Regarding TV sets, a statistical analysis revealed that the product ages of collected waste TV sets were adequately fitted on Weibull distribution (AEHA 2010). Based on this fact, the Weibull distribution was selected for estimating the service life of LCD products in this study. The Weibull probability density function was applied for LCD products shipped at year k and for estimating the waste amount at year t originating from the products shipped at year k. The Weibull probability density function and other settings applied in this study are given as follows:

$$f_i^k(t) = \frac{m_i^k}{\eta_i^k}\left(\frac{t}{\eta_i^k}\right)^{m_i^k-1}\exp\left(-\left(\frac{t}{\eta_i^k}\right)^{m_i^k}\right), \tag{9}$$

$$\mu_i^k = \eta_i^k \cdot \Gamma\left(1+\frac{1}{m_i^k}\right), \tag{10}$$

$$(\sigma_i^k)^2 = (\eta_i^k)^2\left(\Gamma\left(1+\frac{2}{m_i^k}\right) - \Gamma^2\left(1+\frac{1}{m_i^k}\right)\right), \tag{11}$$

Where $f^{\,k}(t)$ is the Weibull probability density function for LCD product i shipped at year k and released at year t (–), $m_i^{\,k}$ is the shape parameter for LCD product i shipped at year k (–), $\eta_i^{\,k}$ is the scale parameter for LCD product i shipped at year k (year), $\mu_i^{\,k}$ is the average lifetime of LCD product i shipped at year k (year), and $\sigma^{\,k}$ is the average standard deviation of LCD product i shipped at year k (year). Using Eq. (5), $\mathrm{W}_i^{\mathrm{LCD}}(t)$ can be calculated as follows:

$$\mathrm{W}_i^{\mathrm{LCD}}(t) = \sum_k^t \mathrm{P}_i^{\mathrm{LCD}}(k) \cdot f_i^k(t), \tag{12}$$

where

$$f_i^k(k) = 0. \tag{13}$$

2.3.3 Evaluation settings

For estimating the future shipment by Eqs. (4–13), available information was collected and combined. The past records of LCD products, i.e., television sets, laptops, and monitors, were obtained from statistics (JEITA 2012; METI 2012a, 2012b; JPIF 2012; JPIA 2012). The shipment forecasting by JEITA was also available (JEITA 2011) and considered in the analysis of material flows, as discussed below. The Weibull parameters were extracted from existing databases on lifespan (NIES 2010; AEHA 2010) (see also Electronic Supplementary Material). The screen size types of laptops and monitors were assumed to be the same ratio as in 2011 (see also Electronic Supplementary Material). Forecasting of the Japanese population was extracted from the report of a national institute on population (IPSS 2012). The ownership ratio of LCD television sets drastically changed in 2009 and 2010, whereas the shipments of laptop computers and LCD monitors in the past several years have not greatly changed. Because of this fact, $\mathrm{OR}_i^{\mathrm{LCD}}(t)$ in Eqs. (5–7) was specified for LCD products, individually. The linear parameters for the growth rate of ownership ratio ($\mathrm{a}_i^{\mathrm{LCD}}(t)$ [(Set product per person) (set product per person)$^{-1}$]): b [(set product per person) (set product per person)$^{-1}$ per year], and c [(set product per person) (set product per person)$^{-1}$] in Eq. (7) were approximated by an extrapolation of the ownership ratio from 2006 to 2011 for laptops and monitors. For television sets, the approximation was based on the extrapolation of the ownership ratio in 2012 and 2013 calculated from the predicted shipment from the industrial association (JEITA 2011).

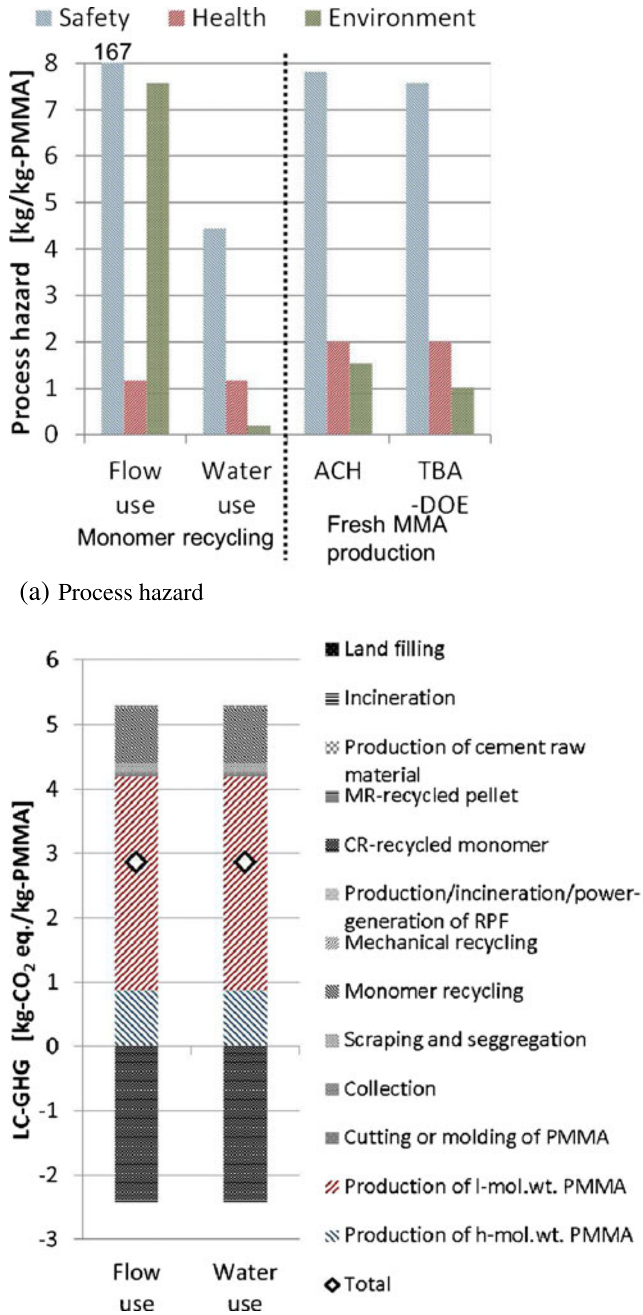
For television sets, the drastic change in product replacement because of the switch to the digital terrestrial broadcasting in Japan on July 24, 2011 makes the estimation complicated. In this study, the minimum shipment in 2012 and 2013 was set as the data from the industrial organization, and the $\mathrm{OR}_i^{\mathrm{LCD}}(t)$ after 2012 was estimated on the basis of the rate in 2012 and 2013. The Weibull parameters were obtained from an investigation report (AEHA 2010), where they were obtained from the records of used home electric appliances collected based on recycling law. In this regard, the average lifetime of collected LCD television sets may have been influenced by the switch to the digital terrestrial broadcasting because the old-type television sets cannot show digital channels, and many people have replaced their television sets, even if the sets have not outlived their usefulness. As for cathode ray tube (CRT) television sets, the lifetime and standard deviation are different from those of LCD sets (see also Electronic Supplementary Material). It can be said that the usage conditions of LCD television sets may become close to those of CRT sets after the broadcasting switch. Considering these points, three scenario sets of Weibull parameters were generated as organized in Table 1 (see also Electronic Supplementary Material). In the first scenario, it is assumed that the usage conditions of LCD television sets are not changed and remain the same as the values in the literature (AEHA 2010). They are changed and are the same as those of CRT television sets at 2010 or 2015 in the second and third scenarios, respectively.

3 Results

Figure 4a shows the process hazards of the two PMMA pyrolysis process options and two fresh MMA production routes that are the most applied routes worldwide and a process with the smallest process hazard according to the literature (Sugiyama et al. 2008b, 2009). In all three hazard categories, the second PMMA pyrolysis process with the water use option has the smallest values. Comparing the two PMMA pyrolysis processes, it is demonstrated that the circulation of cooling water can significantly reduce process hazards. The stream with the highest process hazard was shifted from output flow of condenser to output flow of reactor in Fig. 2a, b, respectively. This is because the MMA monomer has comparatively high hazardous properties and an increased process hazard when a stream contains it in quantity. Their high process hazards originate from several chemicals with hazardous properties treated in fresh MMA production as raw materials, medium-term products, and agents as discussed by Sugiyama et al. (2008b). In this regard, however, the implementation of circulated cooling water slightly increases environmental impacts, as shown in Fig. 4b, and indicated as greenhouse gas (GHG) emission from the PMMA life cycle. The detailed results including process hazards of other MMA production processes are shown in the Electronic Supplementary Material.

The results of the MFA are shown in Figs. 5 and 6. Television sets, laptops, and monitors have individual tendencies; in particular, television sets have a unique tendency (see also Electronic Supplementary Material). In a general trend, the total shipment of LCD panels is linked with the population in Japan. Because the population's demands were mostly saturated in 2011, the shipments of laptops and monitors have similar tendencies as the population decreases. In the records for 2011, the production amount of television sets drastically changed from 2009 to 2011 in Japan, whereas other LCD products have no such peak in the record. This is probably caused by the shift to terrestrial digital broadcasting implemented on July 24, 2011 (MIAC 2011). Because a data converter is needed for television sets to display digital broadcasting, old types of television sets were mostly replaced by new types. This special demand under a rare situation occurred from 2009 to the first half of 2011. The shipment of television sets was considerably reduced in the latter half of 2011. It was shown by the analysis of TV scenarios that such a peak can cause a big fluctuation in shipments.

In TV scenario 1 shown in Fig. 5a, the rapid replacement of released television sets because of the short lifetime of LCD television sets, 5 years, causes much higher production needs in 2012 and 2013 than the industrial association predicted (JEITA 2011). The short lifetime of television sets maintains a large demand for television sets, and the ownership ratio will have also increased (see also Electronic Supplementary Material). This results in a stable production



(a) Process hazard

(b) Environmental impact

Fig. 4 Results of process hazard assessment with the change in environmental impact quantified by LC–GHG based on the same LCA settings as used in previous research (Kikuchi et al. submitted part 1). The results of process hazard assessment of fresh MMA production using the ETH–EHS method from crude oil were obtained from the literature (Sugiyama et al. 2008a), where *ACH* is the acetone cyanohydrin method and *TBA–DOE* is tertiary butyl alcohol (isobutylene) direct oxidative esterification

of television sets, in spite of the decrease in population. TV scenario 2 demonstrates a completely different tendency in shipment, shown in Fig. 5b. The lifetime of television sets reaches the same length as that of CRT television sets, 12.65 years. This means that the shipment around the sharp

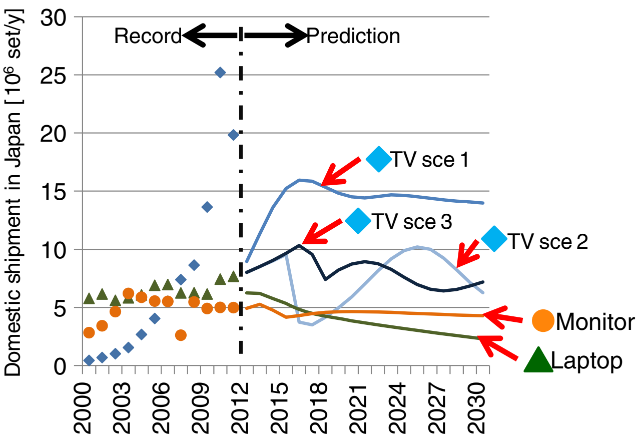


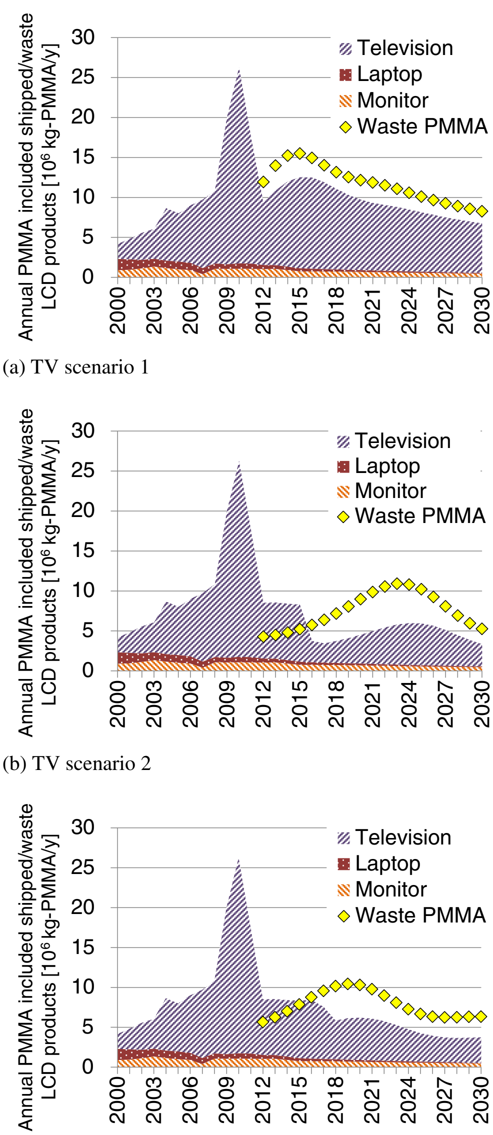
Fig. 5 Predicted domestic shipments of LCD products and lifetime scenarios for television sets

peak in 2010 will be recycled around 2023, and another peak is predicted around 2025. Under the conditions where ownership ratio is not sharply increased, the shipment of television sets greatly decreases around 2016 because the replacement of old type television sets will have ended in a few years, and shipments for replacing released LCD television sets will not become large (see also Electronic Supplementary Material). TV scenario 3 has a similar tendency to TV scenario 2. The lifetime of LCD televisions will gently reach the same length as that of CRT televisions in 2015 rather than in TV scenario 2, which will cause another peak of shipment around 2021. The shipment and waste of PMMA have similar tendencies, as shown in Fig. 6. The shipment and waste for TV scenarios 1 and 2 have peaks after the drastic change caused by the Japanese broadcasting shift. At that time, the amount of PMMA shipment and waste is generally decreased, even though the same number of units is shipped. This is because the contained PMMA in LCDs can be decreased by the promotion of thin-walled processing technology (see also Electronic Supplementary Material).

4 Discussion

The results in this study show the less process hazards of monomer recycling with the circulation of cooling water than the MMA-recycling flow for cooling crude MMA on the meaning of chemical EHS hazards at the early process design phase. It means that the installation of circulation of cooling water leads to inherently safer plant (Kletz and Amyotte 2010). Although the installation of cooling water system may increase some risks on malfunction of cooling system, it may not be largely changed. This is partly because the cooling water system has been already applied in the original process flow, e.g., the cooling of liquefied crude MMA at condenser for adjusting the temperature or the liquefaction of distillate in distillation columns. To analyze in detail the local risks by installation of cooling water system, plant-wide detail risk assessment must be performed in further process design phase because the change in the risk of the installation of cooling water system is shared among other processes in a plant and has relationships with them. At that time, the original process with MMA recycling flow shown in Fig. 2a also must be carefully analyzed.

Although it shows a trade-off relationship between environmental impacts and process hazards, circulation of cooling water could be installed because the increase in environmental impacts is small, and the decrease in process hazards is significant. Such trade-off relationship is discussed more deeply by the weighting of the different types of assessment as organized in Fig. 1. Local risk, such as process hazards, may be prioritized rather than global environmental impacts such as global warming for the decision makers on site. Without any



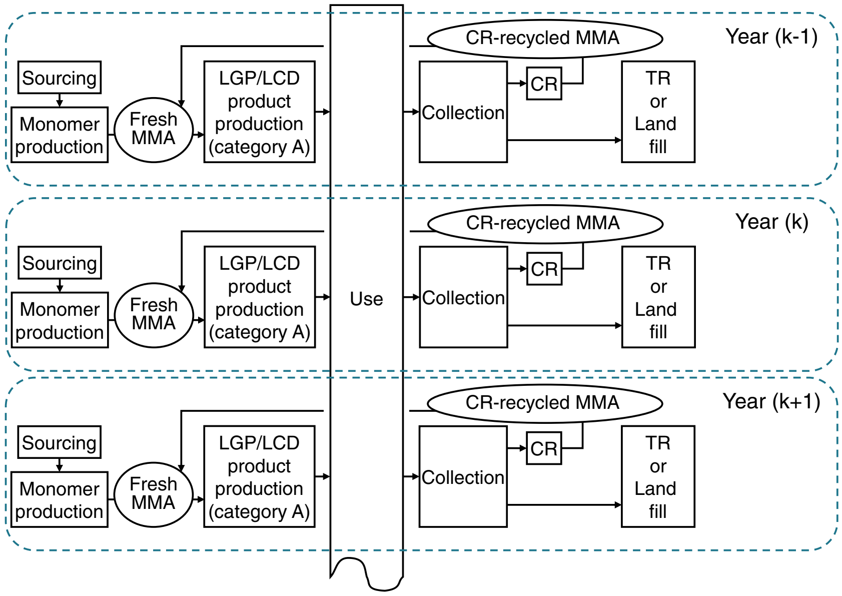
(a) TV scenario 1

(b) TV scenario 2

(c) TV scenario 3

Fig. 6 Predicted PMMA shipment and waste

Fig. 7 Boundary of annual PMMA life cycle



consideration of process hazards, however, a process with the lowest environmental impacts can be attractive. In the case of this study (PMMA monomer recycling), the process hazard can become much higher than fresh MMA production without circulation of cooling water. It might be a critical disadvantage for installing a monomer recycling process. With the circulation of cooling water, the process hazard for monomer recycling is less than that for fresh production of PMMA. This may be partly because the process of PMMA pyrolysis has a simple structure and a reactor for producing MMA, while all fresh MMA production routes have relatively complicated structures and multireactors for producing MMA (Sugiyama et al. 2009).

The collectable waste PMMA in the next two decades can be predicted from the MFA results shown in Fig. 6. The estimated collectable waste PMMA will remain above 5000 t/year in the next two decades in all three TV scenarios. The available waste PMMA from LCD products reached over 10,000 t/year in TV scenarios 2 and 3 around 2023 to 2019, respectively. It decreased and saturated about 5000 t/year during the next 5 years. In TV scenario 1, it reached 15,000 t/year and keeps over 10,000 t/year for about 10 years. To deal with such wide range of collectable PMMA, the recycling system is designed and constructed from the viewpoint of efficiency. To deal with all the available PMMA with the smallest environmental impacts, monomer recycling processes should be applied according to the LCA results in part 1 of this series of papers (Kikuchi et al. 2013, accepted). In this regard, however, the amount of collectable waste PMMA is fluctuated yearly, which means that the operation ratio of constructed process cannot be stable without an adequate storage of wastes. Unstable operation ratio may result in the reduction in the efficiency of processes, and then additional energy is required for recycling unit amount of MMA. Storage, additionally, may require another additional cost and energy. Further analysis is necessitated to check which is better, either monomer recycling with storage for dealing with excess waste PMMA or other general plastic recycling for treating excess waste PMMA. Some general recycling processes such as thermal recycling for PE, PP, and PET has sufficient room in capacity for dealing with excess waste PMMA.

Excess collected PMMA waste over the capacity of monomer recycling may cause a reduction in the price of waste PMMA because the recycling plant of TV set is regulated to operate process to treat waste and recovery materials such as rare earth metal (Yamasue et al. 2009). Waste PMMA contained in them may be regarded as a kind of by-product

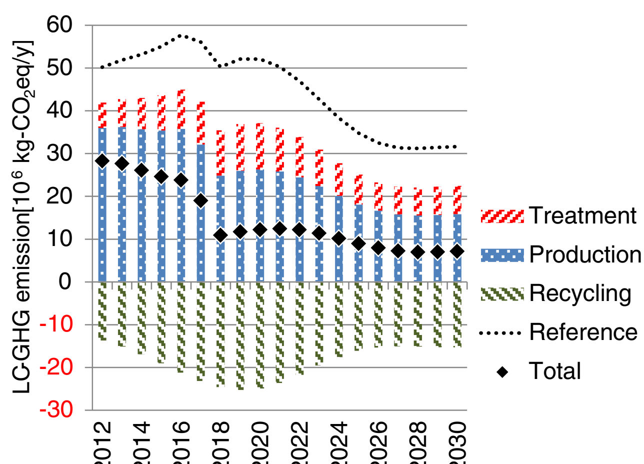


Fig. 8 Result of evaluating annual GHG emission

of home electronic appliance recycling. The cost of monomer recycling was dominated by the procurement of waste PMMA in the actual recycling plant. Hence, the increase of collected waste PMMA in Japan after the shift to terrestrial broadcasting may lead to the decrease of the price of waste PMMA, and thus make a possibility of opening new market of such waste PMMA. At this time, however, according to the LCA of PMMA considering the grade differences in part 1 (Kikuchi et al. 2013, accepted), monomer recycling has an advantage of decreasing the environmental impacts.

At detail process design phase, there should be a discussion of the way to specify the scale of monomer recycling plants: one plant with large throughput or several plants with small throughputs. Distributed plants can decrease the process hazards due to the small amount of stream flow rate. This is also strongly related with the transportation impacts evaluated by LCA because distributed plants require less distance to transport waste LCD products than centralized plant does. In this discussion, it should be taken into account that the larger scale of plant can achieve better efficiency in operation and construction, although it has a risk of decrease in the operation ratio because of the uncertainty of the collectable amount of waste PMMA. The estimated collectable amount of PMMA includes that contained in LCDs collectable at the present according to interviews with a recycling plant for home electrical appliances in Japan, where disassembled LGPs made of PMMA are stockpiled at the present. Because of the existing laws and regulations (METI 2001; PC3R Promotion Association, Japan 2004), LCD panels and their parts can be collected and segregated by manual operations (Hirasawa 1999). The plants were originally brought into practice for collecting other substances, such as rare metals (Yamasue et al. 2009). The PMMA recycling system can apply existing social infrastructure to collect waste PMMA. On the other hand, the monomer recycling process also has a large role in the treatment of production loss from PMMA products production. The plant should be constructed and accept production loss and waste PMMA. This can lead to a robust recycling process that can treat waste PMMA with a stable raw material and production loss.

Based on the result of the MFA, the annual environmental impacts associated with PMMA contained in LCD products can be examined. The boundary and GHG emission are schematically shown in Figs. 7 and 8. The dashed boundary in Fig. 7 shows the annual borderline, which means that one bar graph within Fig. 8 is the GHG emission released in 1 year. The waste PMMA in a year is the cumulative result, as shown in Eq. (12) because of the variety in the time duration of use stage of shipped PMMA. Figure 7 shows the results of TV scenario 3 (see also Electronic Supplementary Material) and a reference scenario where all the produced PMMA is incinerated, including reduction losses and after-use PMMA. It was clearly demonstrated that the PMMA monomer recycling process can reduce total GHG emission in the next decades. According to the MFA result, monomer recycling plants should be constructed by 2016 to keep the total GHG emission at a low level.

Through the three types of assessment, i.e., LCA (Kikuchi et al. 2013, accepted), EHS, and MFA, the PMMA monomer recycling process can be a useful process for circulating MMA with lower environmental impacts than the other existing plastic recycling processes. Although the population is decreasing, and the ownership of electronic devices has mostly reached saturation in Japan, the worldwide demand for LCD panels and PMMA will keep increasing according to Display Search Ltd (2012). There is another possibility that the demand for PMMA can be increased by the replacement of conventional heavy materials, such as glass, with plastics, as discussed by Humbert et al. (2009) and Ribeiro et al. (2006). Demand for shipment of PMMA contained in LCD products automatically causes peaks of waste PMMA. Because the lifetime of television sets might be different from country to country, the plant scale and construction timing must be rechecked specifically for the country conditions. The MFA results in this study can be an example for other countries. Regarding process hazards and environmental impacts, the results of this study can be applicable to support the design process all over the world.

5 Conclusions

The process hazard based on environment, health, and safety categories was evaluated for a PMMA pyrolysis process. It has fewer hazards than does fresh MMA production processes. Based on these results, the PMMA monomer recycling process should be able to be accepted from the viewpoints of environmental friendliness (Kikuchi et al. 2013, accepted) and local risks. MFA results revealed large fluctuations in PMMA shipments and waste because of the demand produced by the shift to terrestrial digital broadcasting in Japan. The results demonstrated that over 5000 t/year waste PMMA will be released in the next decades via the recycling system for home electric appliances and personal computers. Their collectability seems to be high according to interviews with recycling plants for home appliances.

The use of multiple assessment methods can reveal various aspects of technologies. A trade-off relationship between local and global effects was confirmed for PMMA pyrolysis. Although these assessment methods have different aspects in their results, utilized information and data have some similarity, as depicted in Fig. 1. Integrated application of methods, not simply calculating a single index, should be addressed for facilitating the next integrated process/technology assessment, as discussed in the literature (Kikuchi and Hirao 2009).

MFA revealed a fact that a political decision such as the shift to terrestrial digital broadcasting can cause a big change

in the trends of shipment and waste of products, as shown in Figs. 5 and 6. Such a change can sometimes make the development of a recycling system difficult and unstable, and finally result in a collapse of material circulation. To avoid such a high-risk situation, appropriate technology such as PMMA pyrolysis should be examined and installed.

Acknowledgments The authors would like to thank Katsumi Fujisaki (Mitsubishi Electric Co., Ltd. and Hyper Cycle Systems Co., Ltd.) and Tsuruoka Co., Ltd. for their cooperation in the investigation of the recycling processes. The authors also thank Yuma Yuguchi for their cooperation in the simulation of process models and the collection of data for LCA. A productive discussion on MFA with Dr. Ichiro Daigo is appreciated. Part of this study was supported by a grant-in-aid for scientific research (B) (no. 23360404) and scientific research (A) (no. 24246150) from the Japan Society for the Promotion of Science. The author (Yasunori Kikuchi) was supported through the Japan Society for Promotion of Science Institutional Program for Young Researchers Overseas Visits and Young Researchers Overseas Study Program for Mechanical Systems Innovation managed by the Global COE Program, "Global Center of Excellence for Mechanical Systems Innovation," by the Ministry of Education, Culture, Sports, Science, and Technology, Japan.

References

Adu IK, Sugiyama H, Fischer U, Hungerbühler K (2008) Comparison of methods for assessing environmental, health, and safety (EHS) hazards in early phases of chemical process design. Trans IChemE: Part B Proc Saf Environ Protect 86(B2):77–93

Aktas CB, Bilec MM (2012) Service life prediction of residential interior finishes for life cycle assessment. Int J Life Cycle Assess 17:362–371

Arena U, Mastellone ML, Perugini F (2003a) The environmental performance of alternative solid waste management options: a life cycle assessment study. Chem Eng J 96:207–222

Arena U, Mastellone ML, Perugini F (2003b) Life cycle assessment of a plastic packaging recycling system. Int J Life Cycle Assess 8:92–98

Association for Electric Home Appliances, Japan (AEHA) (2010) Investigation on the elapsed years of use for four types of electric home appliances (original title in Japanese). http://www.meti.go.jp/meti_lib/report/2010fy01/E001174.pdf. Accessed 21 May 2012

Center for Chemical Process Safety (2007) Guidelines for risk based process safety. Wiley, New Jersey, USA

Centers for Disease Control and Prevention (CDC) (2012) Documentation for immediately dangerous to life or health concentrations (IDLHs), http://www.cdc.gov/niosh/idlh/intridl4.html. Accessed 21 May 2012

ChemExper Inc. (2012) ChemExper, Catalog of Chemical Suppliers, Physical Characteristics and Search Engine. URL http://www.chemexper.com/. Accessed 21 May 2012

Display Search Ltd. (2012) LCD TV Shipments to grow 7 % in 2012 to 220M units, despite zero growth in overall TV shipments. http://www.displaysearch.com/cps/rde/xchg/displaysearch/hs.xsl/120509_lcd_tv_shipments_to_grow_in_2012_to_220M_units_despite_zero_growth_in_overall_tv_shipments.asp. Accessed 21 May 2012

Environmental Protection Agency (EPA), USA (2011) Estimation Program Interface (EPI) Suite 4.10. http://www.epa.gov/oppt/exposure/pubs/episuite.htm. Accessed 21 May 2012

Fuji Chimera Research Institute, Inc (2010) Market report of plastic sheet. Fuji Chimera Research Institute, Tokyo, Japan

Hirasawa E (1999) A recycling plant for home electric appliances. Mitsubishi Electric Advance 87:7–11

Humbert S, Rossi V, Margni M, Jolliet O, Loerincik Y (2009) Life cycle assessment of two baby food packaging alternatives: glass jars vs. plastic pots, Int J Life Cycle Assess 14:95–106

International Labor Organization (ILO) (2012) The International Chemical Safety Cards (ICSC) database. http://www.ilo.org/dyn/icsc/showcard.home. Accessed 21 May 2012

Japan Electronics and Information Technology Industries Association (JEITA) (2011) Prediction of waste television sets after the sign-off of analog terrestrial broadcasting in Japan, 2011. http://home.jeita.or.jp/page_file/20110624141831_PTQuK26jpg.pdf. Accessed 21 May 2012

Japan Electronics and Information Technology Industries Association (JEITA) (2012) Statistics on domestic shipments of major consumer electronic equipment. http://www.jeita.or.jp/english/stat/index.htm. Accessed 21 May 2012

Japan Environmental Management Association for Industry (JEMAI) (2007) JEMAI-LCA Pro

Japan Petrochemical Industry Association (JPIA) (2012) Statistical data of MMA production, shipment, and stock, http://www.jpca.or.jp/4stat/00index.htm. Accessed 21 May 2012

Japan Plastics Industry Federation (JPIF) (2012) Statistical data of plastic production in 2011, http://www.jpif.gr.jp/3toukei/conts/2011/2011_genryou_c.htm. Accessed 21 May 2012

Jiménez-González C, Constable DJC (eds) (2011) Green chemistry and engineering: a practical design approach. John Wiley & Sons, New Jersey, USA

Kakudate K, Kjikawa Y, Adachi Y, Suzuki T (2002) Calculation model of CO_2 emissions for Japanese cars. Int J Life Cycle Assess 7:85–93

Kaminsky W, Franck J (1991) Monomer recovery by pyrolysis of poly(methyl methacrylate) (PMMA). J Anal Appl Pyrol 19:311–318

Kikuchi Y, Hirao M (2009) Hierarchical activity model for risk-based decision making integrating life cycle and plant-specific risk assessments. J Ind Ecol 13(6):945–964

Kikuchi Y, Hirao M, Ookubo T, Sasaki A (2013) Design of recycling system for poly(methyl methacrylate) (PMMA) Part 1: recycling scenario analysis. Int J Life Cycle Assess. doi:10.1007/s11367-013-0624-y. (accepted)

Kletz TA, Amyotte P (2010) Process plants: a handbook for inherently safer design. CRC Press, Second Edition

Koller G, Fischer U, Hungerbühler K (2000) Assessing safety, health, and environmental impact during early process development. Ind Eng Chem Res 39(4):960–972

Koller G, Fischer U, Hungerbühler K (2001) Comparison of methods suitable for assessing the hazard potential of chemical processes during early design phases. Trans IChemE: Part B Proc Saf Environ Protec 79(B2):157–166

Kolluru RV, Bartell SM, Pitblado RM, Stricoff RS (1996) Risk assessment and management handbook for environmental, health, and safety professionals. McGraw-Hill, New York

Kubo Y (2011) Compression molding machine for thin-walled LCD (original title in Japanese). SangyoKikai 727:11–15

Life Cycle Assessment Society of Japan (JLCA) (2012) JLCA-LCA database 2012 1st edition http://lca-forum.org/database/. Accessed 21 May 2012

Lin CH (2008) A model using home appliance ownership data to evaluate recycling policy performance. Resour Conserv Recycl 52:1322–1328

Liu X, Tanaka M, Matsui Y (2006) Generation amount prediction and material flow analysis of electronic waste: a case study in Beijing, China. Waste Manag Res 24:434–445

Ministry of Economy, Trade, and Industry, Japan (METI) (2001), Home Appliance Recycling Act, http://www.meti.go.jp/policy/recycle/main/english/law/home.html. Accessed 21 May 2012

Ministry of Economy, Trade, and Industry, Japan (METI) (2012a) Paper, printing, plastic, rubber products statistics. http://www.meti.go.jp/statistics/tyo/seidou/result/ichiran/06_kami.html. Accessed 21 May 2012

Ministry of Economy, Trade, and Industry, Japan (METI) (2012b) Statistical survey on trends in industrial production (original title

in Japanese), http://www.meti.go.jp/statistics/tyo/seidou/result/ichiran/03_kikai.html. Accessed 21 May 2012

Ministry of Internal Affairs and Communications, Japan (MIAC) (2011) Consultation center for terrestrial digital television broadcast. http://www.soumu.go.jp/main_sosiki/joho_tsusin/dtv/english/index_en.html. Accessed 21 May 2012

Nakano K, Aoki R, Yagita H, Narita N (2007) Evaluating the reduction in greenhouse gas emissions achieved by the implementation of the household appliance recycling in Japan. Int J Life Cycle Assess 12:289–298

Nakatani J, Fujii M, Moriguchi Y, Hirao M (2010) Life cycle assessment of domestic and transboundary recycling of post-consumer PET bottles. Int J Life Cycle Assess 15:590–597

National Institute for Environmental Studies, Japan (NIES) (2010) Lifespan database for Vehicles, Equipment, and Structures: LiVES. http://www.nies.go.jp/lifespan/index-e.html. Accessed 21 May 2012

National Institute of Population and Social Security Research, Japan (IPSS) (2012) Population Statistics of Japan. http://www.ipss.go.jp/syoushika/tohkei/Mainmenu.asp. Accessed 21 May 2012

PC3R Promotion Association, Japan (2004) House hold PC recycling, http://www.pc3r.jp/e/index.html. Accessed 21 May 2012

Ribeiro C, Ferreira JV, Partidário P (2006) Life cycle assessment of a multi-material car component. Int J Life Cycle Assess 12:336–345

Romero-Hernández O, Romero-Hernández S, Muñoz D, Detta-Silveira E, Palacios-Brun A, Laguna A (2009) Environmental implications and market analysis of soft drink packaging systems in Mexico. A waste management approach. Int J Life Cycle Assess 14:107–113

Scheirs J, Kaminsky W (eds) (2006) Feedstock recycling and pyrolysis of waste plastics: converting waste plastics into diesel and other fuels (Wiley Series in Polymer Science). John Wiley & Sons, Chichester

Sugiyama H (2007) Decision-making framework for chemical process design including different stages of environmental, health and safety (EHS) assessment. Dissertation ETH No. 17186, Swiss Federal Institute of Technology, Zurich

Sugiyama H, Hirao M, Fischer U, Hungerbühler K (2008a) Activity modeling for integrating environmental, health, and safety (EHS) consideration as a new element in industrial chemical process design. J Chem Eng Jpn 41(9):884–897

Sugiyama H, Fischer U, Hungerbühler K, Hirao M (2008b) Decision framework for chemical process design including different stages of environmental, health, and safety assessment. AICHE J 54(4):1037–1053

Sugiyama H, Fischer U, Antonijuana E, Hoffmann VH, Hirao M, Hungerbühler K (2009) How do different process options and evaluation settings affect economic and environmental assessments? A case study on methyl methacrylate (MMA) production processes. Process Saf Environ Prot 87:361–370

Yamasue E, Minamino R, Daigo I, Okumura H, Ishihara KN (2009) Evaluation of total materials requirement for the recycling of elements and materials (urban ore tmr) from end-of-life electric home appliances. Mater Trans 50(9):2165–2172

Yokota K, Matsuno Y, Yamashita M, Adachi Y (2003) Integration of life cycle assessment and population balance model for assessing environmental impacts of product population in a social scale. Case studies for the global warming potential of air conditioners in Japan. Int J Life Cycle Assess 8:129–136